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Comparative Study of Open Source Particulate Emission Measuring Techniques

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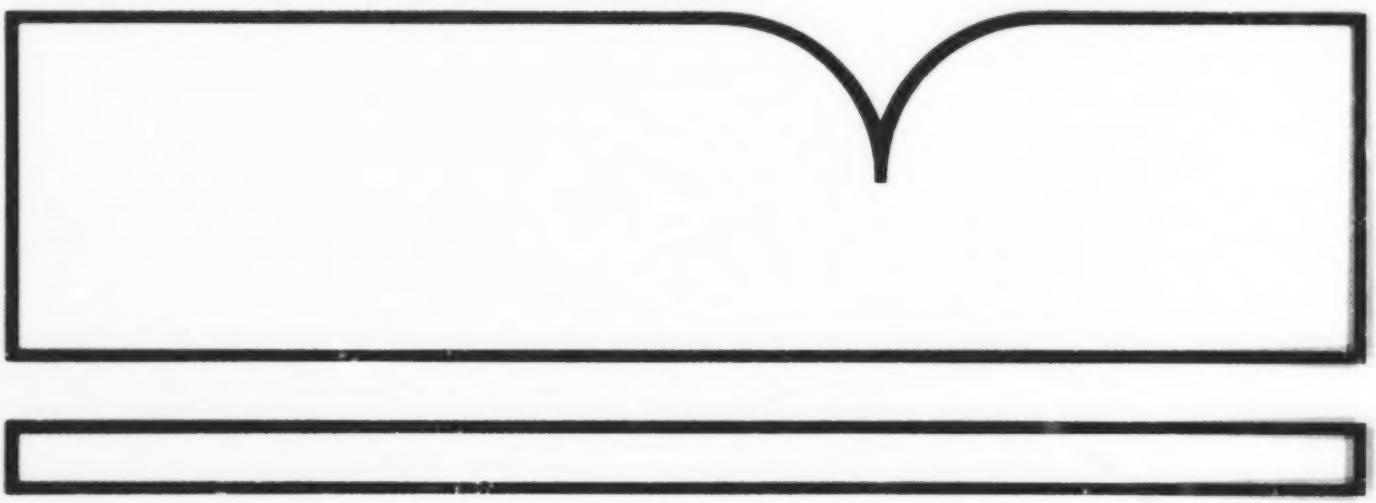

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Comparative Study of Open Source Particulate Emission Measuring Techniques

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16. ABSTRACT The paper gives results of a comparative study of variations in measuring and analytical techniques used to assess sources upon which emission factor data bases are built (in an effort to quantify these technique-dependent parameters). The study included a simultaneous, side-by-side field test with five independent testing organizations sampling dust emissions from an unpaved and uncontrolled road at a major steel producing facility. Each participant independently measured the particulate emission concentrations produced by the roadway traffic. These measurements resulted in not only particle-size dependence of the emissions but also concentrations as functions of the distance above the road surface. The results for each participant were expressed as emission factors for total particulate and the mass fractions of the particulate with size < 30, < 15, < 10, and < 2.5 micrometers diameter. The study showed that all five profiling systems were capable of producing equivalent results in terms of total emissions. This was not the case for emissions by particle size: the only technique found to produce reliable emission factors by particle size was inertial sizing.

17. KEY WORDS AND DOCUMENT ANALYSIS

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Dust	Particulate	14G
Particle Size	Open Sources	
Measurement	Emission Factors	14B
Analyzing		
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ABSTRACT

Current EPA emission trading (bubble) policy allows excessive emissions from one source to be offset by improved controls on other sources within the same industrial complex. In implementing this bubble policy, many industries have opted to reduce particulate emissions from roads, parking lots, and storage locations in lieu of more stringent controls on other plant operations. The predictions of these open sources emissions based upon the existing data for measured emission factors have been found to be subject to high variability. Part of this uncertainty can be attributed to the variations in source characteristics. However, a significant part of the uncertainties are due to variations in the measuring and analytical techniques used to assess sources upon which the emission factor data base was built. In an effort to quantify these technique dependent parameters, a comparative study of the current measuring and data reduction methods was carried out. This study included a simultaneous, side-by-side field test with five independent testing organizations sampling the dust emissions from an unpaved and uncontrolled road at a major steel producing facility. Each of the participants independently measured the particulate emission concentrations produced by roadway traffic. These measurements resulted in not only particle-size dependence of the emissions but also concentrations as functions of the distance above the road surface. The results for each testing organization were expressed as emission factors for total particulate and the mass fractions of the particulate with sizes <30, <15, <10, and <2.5 μm diameter. Based on an analysis of the results, it was found that all five profiling systems were capable of producing equivalent results in terms of total emissions. This was not the case for emissions by particle size. The only technique found to produce reliable emission factors by particle size was the inertial sizing method.

Introduction

During the past decade, research has shown that particulate emissions from open sources such as roads and material storage piles contribute significantly to ambient particulate matter concentrations in many areas.[1] The current EPA emission trading policy allows excessive emissions from one source to be offset by improved control of another source within the same plant. In implementing this bubble policy, many steel plants have agreed to reduce fugitive dust emissions in lieu of tighter controls on process emissions.[2] However, the efforts of several groups to develop equipment and methods for quantifying emissions from nontraditional sources such as roadways have resulted in estimates of emission factors and emission rates with substantial variability. Whereas it is generally agreed that emission factor estimates of process emissions from ducted sources are good to ±50% of a specified measured value, open source fugitive dust emission factors may vary from measured values by an order of magnitude as a result of differences in test methods and a limited data base. In an effort to resolve these differences, the U.S. Environmental Protection Agency contracted with Southern Research Institute (SoRI) to conduct a critical review of the various measurement and predictive methods and to conduct a side-by-side field test in which five testing organizations, whose methods ran the gamut of those which have commonly been applied, would simultaneously sample the emissions from an unpaved road. This paper describes the results of that field test.

Test Description

Because much of the use of the emission trading policy with respect to roadways had been in the iron and steel industry, the desired test site was an unpaved road within an integrated iron and steel facility. Negotiations were undertaken with several companies with regard to possible plant access for the tests; the United States Steel Corporation (USSC) responded favorably, offering the use of their Gary Works. The nature of the tests to be conducted made it necessary to use a moderately long, straight stretch of roadway that was oriented more or less perpendicular to the prevailing wind and clear of local perturbing influences for a length of a few hundred meters. It was also desirable that the road have a moderately high traffic density for emissions generation. Unfortunately, all roads on the property of the Gary Works were either paved or had been treated with chemical dust suppressant. The most suitable road section in terms of physical layout and traffic density was a paved slag haul road parallel and adjacent to Lake Michigan. Consequently, this road was made to simulate an uncontrolled unpaved road by applying a 5-10 cm thick layer of dirt to one 300 m section. This portion of the road was located well away from other potentially confounding sources and wind flow obstructions. Five side-by-side test positions, each approximately 15 m wide, were then laid out on both sides of the road near the center of the simulated unpaved road section. A total of 11 tests were conducted over a 5-day period in June 1984. The participating testing organizations were Energy & Environmental Management, Inc. (EEM), Midwest Research Institute (MRI), PEDCo Environmental, Inc. (PEDCo*), TRC Environmental Consultants, Inc. (TRC), and the United States Steel Corporation's Department of Environmental Affairs (USSC).

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*Company name subsequently changed to PEI Associates, Inc.

Test Methods

All five of the participating organizations utilized variants of the exposure profiling tower system first developed by MRI[3] to measure emission rates. However, there were potentially significant differences in the actual applications of the method with respect to such items as the number and heights of the profiler samplers, the filter orientation within the samplers, the sampling flow rates, the methods used for establishing and maintaining isokinetic sampling conditions, and the methods used to apportion the total emissions among the various particle size fractions. Fractions of interest were TSP (assumed to be the weight fraction smaller than 30 μm), PM10 (the weight fraction contained in particles smaller than 10 μm), and the fine particle fraction (defined as the weight fraction contained in particles smaller than 2.5 μm). (Note: All particle diameters used herein are on an aerodynamic basis unless specifically noted otherwise.) Brief summaries of the methods used by the participants are given in Table I.

The methods used for establishing and maintaining isokinetic sampling conditions were as follows:

EEM - Preset fixed flow rates based on 15-minute average wind speed at a height of 4 m for the period immediately preceding the test. The vertical velocity distribution was assumed to be uniform.

MRI - Periodic manual flow adjustment based on wind speeds from a wind shear power curve fit to measured wind speeds at heights of 1.5 and 4.5 m.

PEDCo - Periodic manual flow adjustments based on wind speed at a height of 4 m with the vertical velocity profiles adjusted for wind shear by means of an assumed power function.

TRC and USSC - Continuous, servo-controlled flow adjustment based on wind speeds measured by thermal anemometers located adjacent to each sampling inlet.

Three distinctly different methods were used for the determination of particle size distributions. Both MRI and PEDCo used forms of inertial sizing, while the remaining three organizations used electron microscopy of the material collected by the profiler samplers.

MRI used a combination of a specially fabricated cyclone for the removal of large particles followed by a Sierra Model 235 high-volume cascade impactor.[4] Two such samplers were used, located adjacent to the profiler tower, one at a height of 1.5 m and the other at 4.5 m. These samplers operated at fixed flow rates of 34 m^3/hr (20 cfm) with intake nozzles of selected sizes such that the samples would be obtained at near isokinetic conditions.

PEDCo used the stacked filter concept developed by Cahill, et al.[5] In the PEDCo implementation of the concept, a stainless steel inlet screen having a 30 μm pore size was used to provide a supposed nominal 30 μm fractionation point. The screen was followed by two Nuclepore filters, the first of which had a pore size (8 μm) selected to provide a 2.5 μm aerodynamic diameter cutoff, while the second, which had a pore size of 0.8 μm, served as the final filter.

EEM, TRC, and USSC all used computer controlled scanning electron microscopy (CCSEM) for measuring particle size distributions.[6] In this method selected portions of the filters from the profilers were removed, placed between finely perforated stainless steel screens, and backwashed with acetone to remove the collected particles. The particle in acetone suspension was then agitated, and the particles were redeposited by vacuum filtration onto 0.2 μm pore size Nuclepore filters. A thin coating of carbon was evaporated onto the redeposited specimen after mounting on SEM sample stubs. The SEM was used in conjunction with an automated image analysis system to detect and determine the dimensions of the projected surface of the particles. Particle volumes were estimated by categorizing the particle's shapes and estimating overall dimensions based on the measurements and assigned category. X-ray fluorescence emissions were used to assign a probable composition class to each particle from which a density was assigned to it. After measuring approximately 1500 particles and assigning estimated volumes and densities to them, a physical size-weight distribution curve was constructed. Aerodynamic diameters were also estimated for each particle based on the assigned category, the measured dimensions, and aerodynamic shape factors.

Uniformity of Test Conditions

One factor of great concern during pretest planning was the possibility of nonuniformity of particle concentrations over the length of the road test section. If the particle concentrations were in fact nonuniform with respect to concentration or size distribution, then comparable results could not have been expected from the various sampling systems, even if they were functionally identical. Any potential effect of nonuniformity of conditions along the test zone was partially obviated by rotating the participants' locations so that each spent 1 day at each of the five test positions. The lack of a reference standard for particle size distribution measurement made it difficult to verify uniformity of size distribution; however, uniformity of concentration was checked by means of a conventional high-vol sampler at each test location and by comparison of concentrations calculated directly from the various profiler results.

Bar graphs of the concentrations by test and location are shown in Figure 1. The data shown in the figure revealed no systematic bias with respect to sampling position. The overall average concentration for all tests at each test position ranged from 11.0 to 15 mg/m^3. The differences were not statistically significant from the grand average of 13.3 mg/m^3 at the 90% confidence level. Average concentrations at each position at a height of 2 m as measured by the profilers ranged from 11.7 to 17.4 mg/m^3 with a grand average of 13.0 mg/m^3. Again, statistical tests indicated that no significant positional biases existed. In comparing the hi-vol data from each position with the others, the correlation coefficients, with one exception, ranged from 0.52 to 0.97 indicating that the test to test concentration variations were reasonably uniform along the test strip.

Total Particulate Emission Factors

Of the five participants, four purportedly measure total particulate emission factors while the fifth, PEDCo, purportedly measures only the fraction nominally smaller than 30 μm. Average total emission factors for the complete

test series for the four contractors whose methods normally provide them, were 8.34, 8.25, 8.41, and 5.73 kg/VkmT. The last value quoted, 5.73 kg/VkmT, is significantly different from the other three values at the 90% confidence level. TRC, whose data produced that result, stated in their report that their values were generated from profiler filter catches only and did not include nozzle and inlet washes. Although the omission of the latter material could explain the low values, USSC, using a virtually identical profiling system, also did not include nozzle and inlet losses, and their results produced the highest of the four averages, making the omission of the washes by TRC less likely as the explanation of their low values. A histogram of the total emission factor results from the four contractors who normally report them is shown in Figure 2. In general, the values reported are well correlated with correlation coefficients among the four sets of data ranging from 0.584 to 0.802.

The inlet nozzles and screens of PEDCo's profiler were washed in three of the tests (8, 9, 10) to provide a limited basis of comparison of total emissions with those of the remaining contractors. For those tests, PEDCo's total emission factors averaged 5.8 kg/VKmT while those of the others averaged 6.2, 5.9, 4.6, and 5.1 kg/VKmT, respectively. Thus there is no reason to believe that PEDCo's system would produce values which differed from those of the other contractors if total emission factors were desired.

Particle Size Distribution Results

Three fundamentally different particle sizing methods were utilized by the test participants. USSC, TRC, and EEM used Computer Controlled Scanning Electron Microscopy (CCSEM), MRI used inertial particle sizing devices, and PEDCo used a combination of forms of sieving and inertial devices. In addition, SoRI operated collocated inertial sizing samplers at selected locations during some of the tests. The PEDCo profiling system provided data in two size fractions at each profiler level for each test. MRI's system provided data for each test in seven size fractions at each of two heights for a location adjacent to their profiler. The remaining three participants used CCSEM analysis of varying numbers of the filters from their profilers. USSC performed size analyses on all of the filters from their profiler. TRC had CCSEM analyses performed on two or three filters from selected heights from one test each day. EEM had analyses performed on complete profiler filter sets from two tests which were selected as representative of typical conditions through the week. The analyses for both TRC and EEM were performed by Energy Technology Consultants (ETC) of Murraysville, PA. USSC performed their CCSEM analyses in-house using methods identical to those of ETC.

The results from disparate methods were not found to be mutually consistent. Comparative particle size distribution results from one test are shown in Figure 3, in which marked differences can be seen to exist among the results from the various methods. These differences and their probable causes will be discussed in detail in the following paragraphs.

Averages and standard deviations of emissions factors for total particulates, TSP size range particulates (<30 μm), PM10 (<10 μm), and fine particles (<2.5 μm) for each participating organization are given in Table II. In general, at the 95% confidence level, no significant differences are found in the total emission factors other than the low values from TRC. In the case

of the TSP emission factors, the results from the three contractors using CCSEM analysis show no differences between them which are significant at the 95% confidence level. However, the MRI results are significantly lower than those of the CCSEM users, and the PEDCo results are lower than those of MRI at the 95% confidence level. PM10 emission factors did not differ significantly among USSC, TRC, EEM, and MRI; however, the PEDCo results were again systematically low throughout the tests. Finally, in the fine particle fraction the MRI results were systematically high as compared to those of the other participants at the 95% confidence level.

Looking first at PEDCo's results, they were systematically low for all particle sizes considered. PEDCo's method provides data only at nominal sizes of 2.5 and 30 μm from which 10 μm values are inferred by interpolation. Thus if both the 2.5 and 30 μm data are low, then it is highly likely that the 10 μm value will be as well. It is concluded that the 2.5 and 30 μm values were low, based on comparisons between (1) the ratio of TSP to hi-vol concentrations and (2) the percent of the total PEDCo profiler catch penetrating the screen as a function of total sampler catch.

Addressing comparison (1), it will be shown later that data from other participants indicated that the particle size distribution remained fairly constant at heights between 1.5 and 4.5 m throughout the test series, leading to the expectation that the ratio of TSP concentration to total concentration would be constant as well. The particulate concentrations obtained from the hi-vols were previously shown to be consistent with those measured by the profilers at a height of 2 m. Since the profiler and the hi-vol sampling periods were identical, the hi-vol filter weights can be used as surrogates proportional to the missing total particulate weights for the PEDCo profiler samples at the 2 m level, and the hi-vol concentrations as surrogates for the total concentrations that would have been obtained by the profiler. The points plotted in Figure 4a are the ratios of the TSP (<30 μm) concentrations at the 2 m height from PEDCo's profiler to those of the collocated hi-vols plotted versus the corresponding hi-vol catch weight. As can be seen the ratio declines as the filter weight, and hence the expected total catch weight of the profiler, increases.

The percent of the total PEDCo profiler catches penetrating the screen was determined by analyzing the results from those tests where PEDCo recovered the entire catch. Washes of the sampler screens and inlets were obtained in each of tests 8, 9, and 10. Consequently complete data on the system catches were available for those tests. The ratios of apparent TSP to total concentrations from the lower samplers of PEDCo's profiler, where the distribution might be expected to be fairly constant, are shown in a similar way for tests 8, 9, and 10 in Figure 4b. Again, the ratios tended to decline as the catch weights increased which is indicative of decreasing screen penetration as the profiler samplers loaded up during a sampling run.

The combined effects of two mechanisms are postulated as explaining the low weights obtained by PEDCo. The mechanisms are filtration and misassignment of the screen's particle size cutoff. The physical mechanism used to provide the 30 μm size cut in the PEDCo system is a wire mesh screen with 30 μm openings. The assumption is then made that this inlet screen will provide a particle size cutoff of 30 μm. However, it is probable that significant

collection of particles smaller than 30 μm will take place by impaction and interception even with clean screens, thereby reducing the effective cutoff diameter to a value substantially smaller than 30 μm in much the same way that the 8 μm pores of the Nuclepore filter produce an inertial separation near 2.5 μm. Further, as particles are collected by the screen, the openings are reduced in size making the screen become a progressively more efficient filter. This filtration effect will then result in depressed values for particle concentrations and emission factors for all particle sizes that are assumed to be passed by the filter. (It is worth noting that the openings in the weaves of fabric filters used for particulate control devices are comparable to or larger than the 30 μm openings used here.) Figure 4a, which is based upon the PEDCo data obtained in this test series, illustrates that the screens undoubtedly do become more efficient filters as a test proceeds.

The filtration effect, together with the likely misassignment of the screen's particle size cutoff, is the probable explanation of PEDCo's particle size distribution curve shown in Figure 3 falling below those of the other participants. Even in the absence of these problems, the PEDCo system can provide, at best, questionable data with respect to PM10 emissions. The system is purported to provide size distribution information at only two particle sizes, 2.5 and 30 μm, both of which are relatively far removed from 10 μm. Thus the 10 μm fraction must be estimated by means of some assumptions regarding the shape of the distribution at each height. Even the most commonly used distribution form, the log-normal approximation, cannot be used in PEDCo's normal practice as the total concentrations are not usually determined but only the concentrations of particles nominally smaller than 2.5 and 30 μm are measured.

Turning now to the CCSEM method, Figure 5 shows a particle size distribution obtained by EEM/ETC using CCSEM methodology and the distribution measured by SoRI using a collocated Lundgren cascade impactor. Six such collocated sampling runs were obtained, and all showed results like those illustrated in the figure. The points shown for the impactor results for sizes smaller than 15 μm are from gravimetric determinations. The impactor results for the larger sizes were obtained by measuring the size distribution of the particles collected on the first stage of the impactor by means of optical microscopy and apportioning the mass on that stage accordingly. A total of nine valid downwind Lundgren impactor runs were made over the course of the tests, cursory examination of which revealed that all had collected particles with sizes up to 300 μm, six had particles larger than 400 μm, and two had particles present which were larger than 500 μm. Similarly, MRI reported particles as large as 300 μm in their cyclone catches. No particles physically larger than 80 μm were reported in the data from any of the three groups using CCSEM for size distribution measurement, and in the case of TRC none larger than 50 μm were reported. USSC states that the particle sizing range capability of their CCSEM system is 0.2 to 80 μm, and it appears from the results reported by ETC (EEM and TRC) that the same upper limit applies in practice to their method as well. The omission of the particles in the size classes larger than 50 or 80 μm obviously introduces a bias into the final reported distribution, the severity of which depends on the relative fraction of the mass actually contained in that fraction. In the present instance as much as 25 to 50 percent of the total mass was probably contained in particles physically larger than 80 μm, which mass would be reassigned to smaller particles in the CCSEM results.

Thus, the reported CCSEM distributions would rise much more steeply than the true distributions.

At the other end of the size spectrum, it is unlikely that the techniques used to remove the particle samples from the filters would be effective for removing individual small particles and that the bulk of the small particles removed would be in the form of agglomerates which would be counted as a lesser number of larger particles. This would have the effect of transferring mass from the fine particle end of the distribution to the middle range thereby depressing the fine particle tail of the distribution and again steepening the apparent distribution.

Thus aside from the difficulties in estimating particle volumes and assigning aerodynamic diameters to irregular dissimilar particles of the types encountered here, there appear to be fundamental difficulties at both the large and small ends of the size spectrum that render the CCSEM methodology unsuitable for applications of this type.

One of the primary advantages attributed to PEDCo's system and to the CCSEM particle sizing methodology is that the methods permit the particle size determinations to be made directly on the profiler samples. Thus the size distributions can be obtained at each height from a sample which was acquired isokinetically. The MRI methodology on the other hand provides data obtained directly on an aerodynamic basis using well characterized and widely accepted techniques, inertial separation by cyclones and cascade impactors; and the particle size cuts span the range over which data are needed with adequate resolution. However, their data are obtained at only two heights (1.5 and 3.5 m) from samples that are taken under only quasi-isokinetic conditions. The latter should constitute no real problem except for tests conducted under conditions of widely varying wind velocity. With respect to the former, it would be desirable to have particle size data for at least one more height, say about 7 m.

Conclusions

The five profiling systems were found to be capable of producing equivalent results in terms of total emissions. This was not the case for particle size distributions and emissions by particle size. The lack of an objective standard for particle size distribution determination obviously limits the validity of critiques of methods as needed here. However, long recognized problems in reconstituting size distributions of airborne particles from resuspensions of collected bulk material lead to the conclusion that for fugitive emission sampling of the type undertaken here the sizing should take place prior to collection (or concurrent with collection as in cyclones and impactors). And because of the probable filtration effect and lack of size resolution near 18 μm, the screen inlet/Nuclepore filter method cannot be recommended. Therefore, the only recommended sizing method is that of the cascade impactor which need not necessarily be that used by MRI, although it appears to be acceptable.

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Table I. Profiling system characteristics.

	Organization				
	EEM	MRI	PEDCo	TRC	USSC
No. of heights	5	5	4	5	4
Minimum height (m)	2	1.5	1	1	2
Maximum height (m)	10	7.5	9	9	9.5
Flow control	manual, fixed	manual, variable	manual, variable	automatic	automatic
Sampler flow set by wind speed at (m):	4	1.5 & 4.5	3	each sampler	each sampler
Particle size method	CCSEM[a]	cyclone/ impactor	stacked filter	CCSEM	CCSEM
No. of heights for particle size	3	2	4	3	4
Sampling rate (m^3/hr)	34	34	6.8	34	34

[a]Computer controlled scanning electron microscopy.

Table II. Average emission factors by size class.

	Emission factor, kg/V Km T							
	Total		<30 μm		<10 μm		<2.5 μm	
	Average	σ	Average	σ	Average	σ	Average	σ
EEM	8.3	2.5	6.6	2.0	1.9	0.63	0.21	0.10
TRC	5.7	1.4	5.3	1.3	1.8	0.45	0.16	0.04
USSC	8.4	3.4	7.1	3.2	2.6	1.10	0.32	0.21
MRI	8.3	3.8	3.6	1.7	1.9	0.88	0.61	0.28
PEDCo	5.8[a]	2.0	2.4	0.99	1.2	0.53	0.30	0.21

[a]Three tests only.

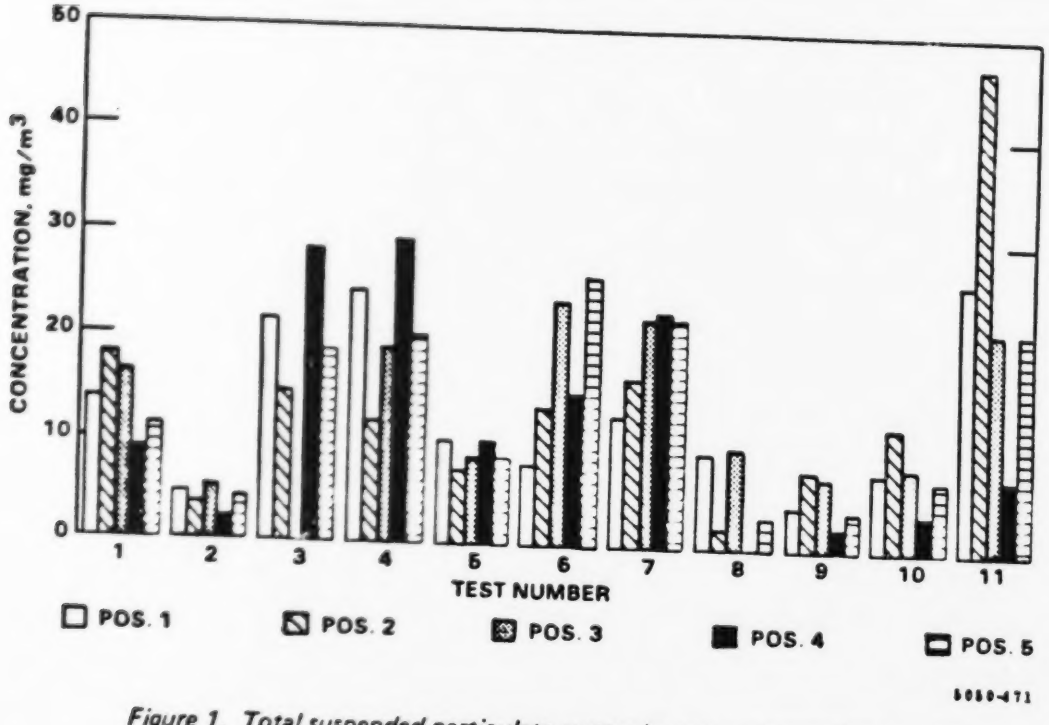

Figure 1. Total suspended particulate matter by test and sampling position as measured by standard hi-vol samplers.

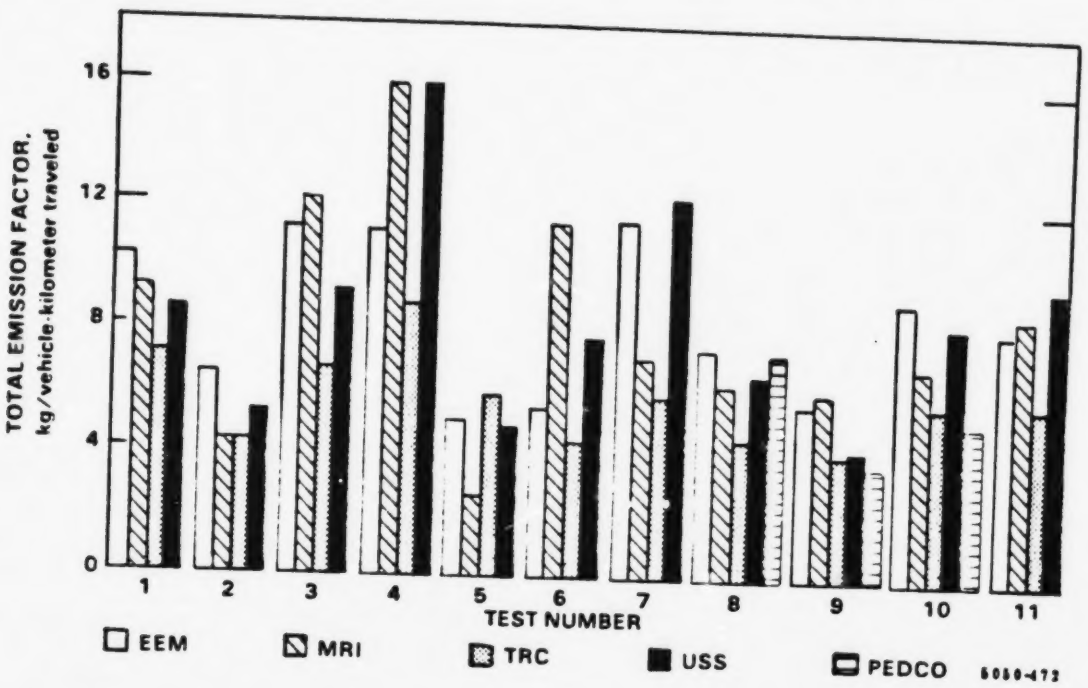

Figure 2. Total particulate emission factors by test as measured by each organization.

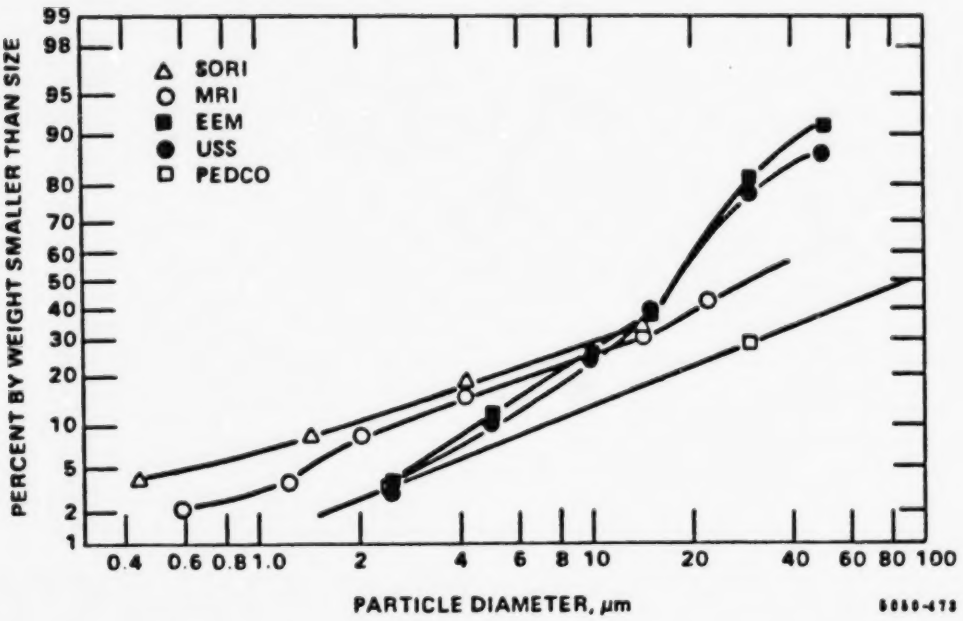

Figure 3. Particle size distributions as measured by the various techniques.

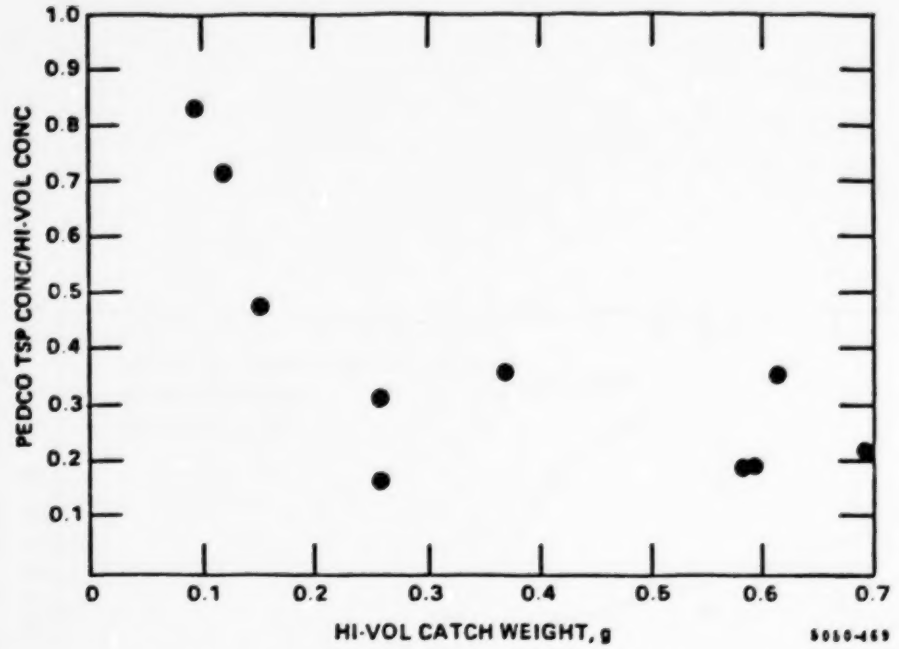

Figure 4a. Plot of the ratio of PEDCO's TSP concentration to hi-vol concentration versus the total particulate mass collected by the hi-vol.

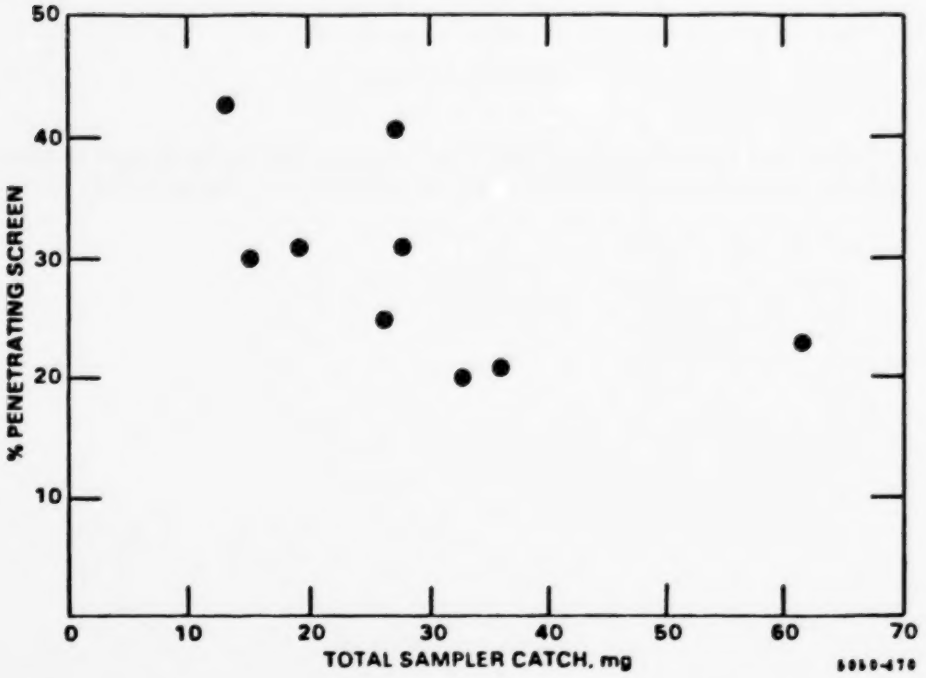

Figure 4b. PEDCO screen penetration dependence on total mass collected by the profiler sampler.

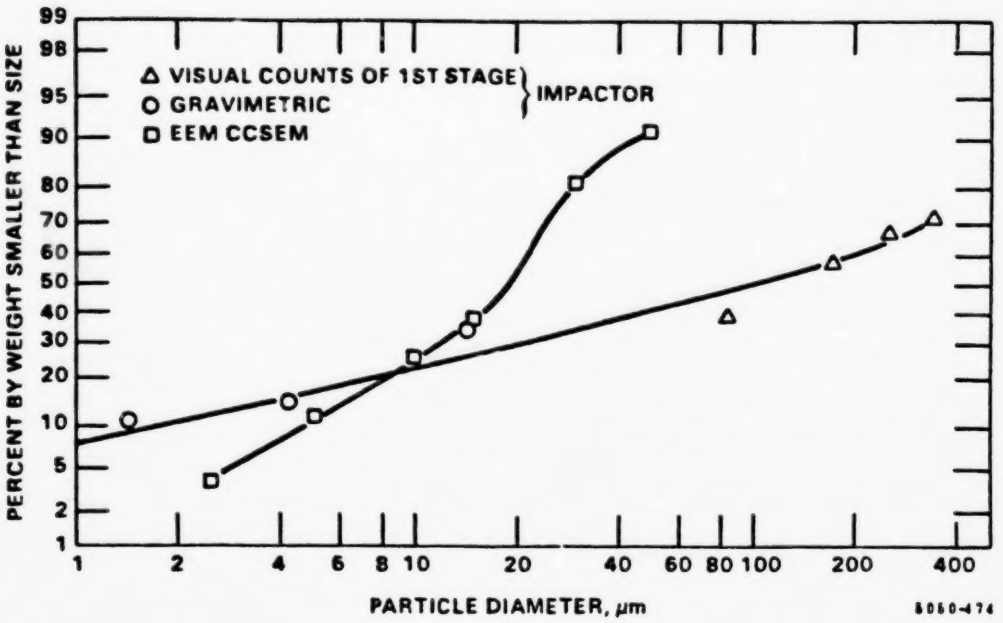

Figure 5. Particle size distribution as measured by a Lundgren cascade impactor and as determined by the CCSEM technique from a collocated profiler sample.